Center for Electrical Energy Storage EFRC

Center for Electrical Energy Storage (CEES): Tailored Interfaces

The Center's overall mission is to acquire a fundamental understanding of interfacial phenomena controlling electrochemical processes that will lead to a dramatic improvement in the performance of electrochemical energy storage devices, notably batteries and supercapacitors.

Batteries and electrochemical energy storage are central to any future alternative energy scenario. Future energy generation sources are likely to be intermittent, requiring storage capacity during inactive times. Batteries are the likely long-term storage solution of choice. The growing reliance on lithium batteries will continue for consumer electronics, aerospace, defense, telecommunications, and medical applications in the near term. In the longer term, we will need them for stationary energy storage for uninterrupted power supply units, the electrical grid, and transportation. Of all systems, rechargeable lithium batteries offer the greatest chance for breakthrough opportunities, and in time, these batteries are destined to constitute a "lithium economy."

While lithium-ion batteries have been successfully implemented in relatively small devices such as cell phones, laptop computers, and cordless power tools, the entry of this technology in heavy-duty systems such as hybrid electric vehicles (HEVs) and plug-in hybrid electric vehicles (PHEVs) has been slowed by barriers relating to calendar and operating life, safety, and cost. The performance limitations arise largely because of uncontrolled reactions that occur at high and low potentials at the electrolyte/electrode interface, leading to high cell impedance, reduced energy and power output, and a limited cycle life (less than two years).

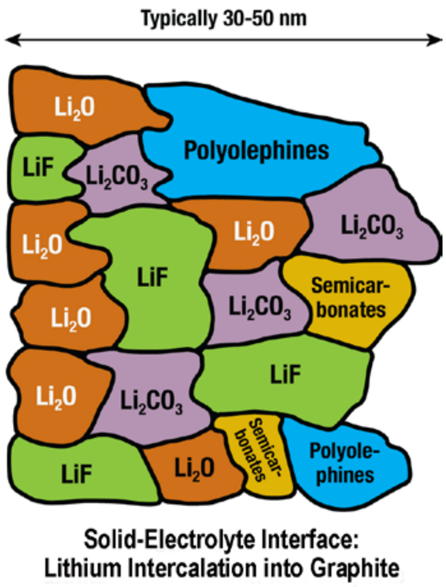
While electrode/electrolyte interfaces and interfacial processes constitute weak links in all electrochemical energy storage devices, these corrosive reactions are not always detrimental to the operation of batteries and supercapacitors. Such reactions can also act positively to create passivating, protective layers that allow rechargeable reactions to occur repeatedly over many electrochemical cycles.

Control and understanding of the composition and structure of electrified interfaces, which is core to the mission of this Center, are essential to overcoming present-day limitations and providing the fundamental basis for finding breakthrough technologies for the next generation of electrochemical energy storage devices and beyond. Success in this endeavor will allow the design of a new generation of materials that can operate safely at high and low potentials and provide, uncompromisingly, the necessary increases in energy and power to enable an improved fuel economy and the emission benefits of HEV and PHEVs, and a reduction of the nation's dependence on foreign oil.

Previous work has emphasized the role of the solid-electrolyte interfaces (SEIs) and interphases (collectively, the SEI layer) as critical components in electrochemical energy storage. An SEI forms in response to the thermodynamic instability of the electrode-electrolyte interface, creating a complex heterogeneous three-dimensional collection of secondary phases and insulating layers having many solid-liquid and solid-solid interfaces. SEI layers have dynamic evolving structures characterized by transverse and longitudinal heterogeneities and compositional and structural gradients. The dynamic creation of SEI layers at electrode-electrolyte interfaces by complex potential-dependent and concentration-dependent processes leads to a weakened, defect-laden structure that is the singular factor limiting the safety, performance, and capacity of present-day battery constructs.

The Center is organized around three individual but strongly interconnected tasks in electrochemical energy storage that address:

- Common issues of electron transfer,



The solid-electrolyte interface is a critical component in electrochemical energy storage.

- Dynamics of cation and anion transfer at the electrode-electrolyte interface, and
- The interplay of materials and architectures at all length and time scales.

Three-Dimensional Architectures at the Electrode/Electrolyte Interface

This task focuses on the design and electrochemical evaluation of three-dimensional electrode/electrolyte interfaces using novel scaffolds, nano-architectures, and surface structures. The task includes experimental studies and theoretical modeling of anode/electrolyte and cathode/electrolyte interfaces.

Dynamically Responsive Interfaces

In this task, the focus is on microcapsules and electrolyte additives to improve battery safety and longevity.

Major activities include:

- Engineering of microcapsule shell walls to protect core contents and release core contents with an appropriate triggering mechanism;
- Development of encapsulated phases for electrode shutdown (battery protection) and damaged electrode restoration;
- Theory and modeling to design and select suitable microcapsule and electrolyte additives; and
- Testing of functional responses of microcapsules, healing agents, and electrolyte additives, including redox shuttles and those forming stable passivating layers on the electrodes.

Understanding and Control of Interfacial Processes Relevant to the SEI

The central focus of this task is the characterization of SEI layers and architectures relevant to the processes that limit the performance of energy storage materials, such as materials strain caused by lattice expansion due to Li incorporation, the role of lateral heterogeneities, the breakdown of solvent molecules at elevated potentials, and the role of additives in stabilizing the interface. The initial focus will be to develop an in-depth understanding of surface structures by in-situ characterization, processes associated with SEI structures and their formation, and the role of passivating layers in extending materials' performance.

The Center brings together a world-class team of 17 scientists from Argonne National Laboratory, the University of Illinois at Urbana-Champaign (UIUC), and Northwestern University (NU). All organizations will leverage Department of Energy user facilities at Argonne: the Advanced Photon Source, the Center for Nanoscale Materials, the Electron Microscopy Center for Materials Research, and the Argonne Leadership Computing Facility. Facilities at UIUC include the Center for the Microanalysis of Materials and the School of Chemical Sciences Facilities; NU's facilities include the Nanoscale Characterization and Experimental Center.

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